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Mitigating the Effects of Quaternary Ammonium Compounds on Biological Wastewater Treatment Systems during the COVID-19 Pandemic

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Abstract

The global pandemic of COVID-19 has sparked an increase in disinfectants' usage around the world. Among these are quaternary ammonium compounds (QACs), which are found in cleaner formulations that are commonly used in various sanitation applications. QACs' concentrations are projected to increase in the influents of wastewater-treatment plants, where their poor target specificity will, initially, adversely affect the plant performance. This paper provides an insight into the possible mitigation of QACs' effects on the treatment plants by manipulating a group of the plant operational parameters (each individually), to alter the plant operation in order to combat these effects. The study used computer simulations of a benchmark model for a treatment plant that take into consideration reactions taking place in the biological units (Anoxic and Aerobic). It was found that increasing the solids' recycle ratio (up to 4), reducing the influent wastewater (by a fraction no less than 0.8), and increasing the oxygen supply rate to its maximum are effective measures that will alleviate the effects of QACs on the plant performance. Altering the nitrate recycle ratio had no tangible effects. Results presented here should be helpful in maintaining an adequate operation of the plants during the first exposure to increasing QACs' concentration, while the model can provide a framework for assessing wastewater treatment plants' performance upon receiving inhibitory material in their influents.

Keywords: COVID-19, Quaternary ammonium compounds, Inhibition, Mitigation, Modeling.

1. Introduction

In February 2020, the World Health Organization (WHO) officially named the disease caused by the novel corona virus as Corona virus Disease 2019 (COVID-19) (Zu et al., 2020), and in March of the same year, the WHO declared the COVID-19 outbreak a global pandemic (Cucinotta and Vanelli, 2020). The spread of COVID-19 triggered the WHO to publish a multitude of guidelines to ensure the prevention and control of this epidemic (WHO, 2020), which include the disinfection of hard surfaces, as they provide a medium for the transmission of the virus (WHO, 2020; Thomas et al., 2020; Van Doremalen et al., 2020). One of the recommended disinfectants for this purpose are virucidal agents that contain Quaternary Ammonium Compounds (QACs) (EPA, 2020; Marnie and Peters, 2020). QACs represent well-known disinfectants composed of a hydrophobic hydrocarbon chain (one at least) connected to a nitrogen atom (positively charged), and another short-chain alkyl group such as methyl or benzyl groups. They are effective in eliminating the viral infection by targeting the lipid structure of the viral envelope (Cross and Singer, 1994). In general, QACs are extensively used worldwide, where their annual consumption was estimated in 2004 to be half a million ton (Chen et al., 2018). Furthermore, more than one million pounds of QACs are manufactured or imported annually, designating them as "high production volume chemicals" by the EPA and the Organization for Economic Cooperation and Development (Hora et al., 2020). The extensive usage in domestic applications (among many others) has led to the detection of QACs in many Wastewater Treatment Plants' (WWTP) influents, and treated effluents, as well as in surface water, and sediments. Concentrations of QACs were detected to be between 1 and 60 μg/l in effluents and surface water (Ding and Liao, 2001; Ferrer and Furlong, 2001; Martínez-Carballo et al., 2007; Tezel and Pavlostathis 2011; Zhang et al., 2015; Pati and Arnold, 2020), and at concentrations ranging from 0.3 to 3 mg/l in WWTP influents (Conidi et al., 2019). Additionally, QACs were found in samples collected from sewers at concentrations ranging between 4.7 and 7.7 μg/l (Wieck et al., 2018), while sewage sludge was found to contain QACs at concentrations ranging between 0.09 and 191 μg/g of dray weight (Ruan et al., 2014). In the environment, QACs are known to proliferate antibiotic resistance in microbial communities (Pereira and Tagkopoulos, 2019), while being toxic to aquatic and soil organisms (Jardak et al., 2016). Additionally, QACs are known to form a carcinogen known as N-Nitrosodimethylamine (NDMA), when combined with chloramines (Kemper et al., 2010). Chloramines results from chlorine disinfection, in the presence of the ammonia, of the effluents of WWTPs prior to their release into the environment (Mitch and Sedlak, 2004).

QACs inhibit the operation of WWTP by targeting the biologically driven reactions. The WHO clearly states that

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there is no evidence of COVID-19 transmission through wastewater, within the confinement of WWTP (WHO, 2020). However, as an indirect consequence to the global pandemic, WWTPs operation could be negatively affected. As a response to the global COVID-19 pandemic, an expected rise in the usage of QAC-containing disinfectants is projected in various locations to combat the spread of the disease. Accordingly, and in an indirect way, QACs' concentration in WWTP influents will increase to higher levels than reported before.

This, by its turn, will present a challenge to WWTP operators due to the adverse effects of QAC-containing wastewaters on the biological treatment systems in such plants. QACs are highly effective disinfectants, yet they have poor target specificity, attacking the microbial group operating within biological WWTPs. Their inhibitory effect to respiratory enzymes will result in a decreased chemical oxygen demand (COD) and the substrate utilization rates (Zhang et al., 2011; Hajaya and Pavlostathis, 2012). QACs will cause total inhibition of nitrification at concentration of 2 mg/l (Sütterlin et al., 2008; Hajaya and Pavlostathis, 2012), and a QAC concentration greater than 50 mg/l has been reported to inhibit heterotrophic denitrification (Hajaya et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015).

The fate and effect of QACs in WWTPs will mainly be dictated by three processes: adsorption, inhibition, and biotransformation (Hajaya and Pavlostathis, 2012; Chen et al., 2018). The aforementioned processes represent how WWTPs interact with QACs in their influents under conventional circumstances. However, an expected increase in usage due to the unusual circumstances brought by the global pandemic (Tullo and Bettenhausen, 2020) represents a new challenge for WWTPs, as a great percentage of the compounds will ultimately find way into their influents (Tezel and Pavlostathis, 2011).

Ultimately, WWTPs will cope with the presence of QACs in their influent wastewaters, with the effectiveness of the process reaching pre-QACs exposure levels through microbial acclimation, biodegradation, or adsorption (Ren et al., 2011; Hajaya and Pavlostathis, 2012; Chen et al., 2018), but not before going through an operational period that will drastically affect the plant's nitrogen removal efficiency, resulting in high concentrations of nitrogen in the plant effluents during that period (Hajaya and Pavlostathis, 2012).

Consequently, there should be criteria within the operational parameters available to the operators of WWTPs to allow them to cope with the expected and temporary decline in their performance while treating wastewaters that include higher levels of QACs (Conidi et al., 2019).

The purpose of this work is to provide an insight into the range of operational parameters that can be exploited to mitigate the impact of a sudden increase of QAC concentrations in the influent domestic wastewater of a WWTP, because of their extensive usage to combat the spread of COVID-19 infection. This task will be performed through mathematical simulations of a benchmark WWTP model that takes into account the inhibitory effect of QACs on the various biological processes in the WWTP.

2. Methods

2.1 The Layout of the Benchmark WWTP

The WWTP model mainly adapted the characteristics of the BSM1 benchmark (Alex et al., 2008). Figure 1 shows the plant layout.

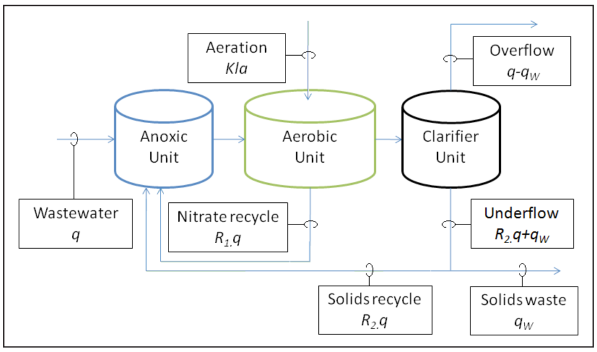

Figure 1. The WWTP layout used in the mathematical model simulations.

Similar to the benchmark, the WWTP is sized for an average domestic wastewater flow rate (q) of 18500m^3/d. The organic load in the wastewater is comprised from 300 mg COD/l of biodegradable organic carbon along with 50 mg N/l of nitrogen. The anoxic and aerobic unit volumes are 2000 m^3 (V_1) and 4000 m^3 (V_2), respectively. Two recycles are found: a nitrate recycle (q_{R1}) with a recycle ratio of 5 (R_1), and a solids' recycle (q_{R2}) with a recycle ratio of 1 (R_2). These ratios dictate the value of the nitrate recycle flow rate ($q_{R1}= R_1.\ q$) and the solids' recycle flow rate ($q_{R2}= R_2.\ q$) in relation to the flow rate of the wastewater.

Solids' waste flow rate (q_w) is 385 m^3/d, in order to sustain a Solids' Retention Time (SRT) of about sixteen days. Air is only introduced into the aerobic unit to sustain an oxygen concentration of around 2 mg/l. The aforementioned oxygen concentration is reached by assigning a value for the oxygen mass transfer coefficient (Kla) of 200 (1/d).

The clarifier sub-model is not included in the model. Instead, it is assumed that no reactions are taking place in it, and its particulates' separation efficiency (SE) is relatively constant (Le Moullec et al., 2011; Hajaya, 2019). SE was calculated from the benchmark model to be 0.998 (particulate mass in the underflow/ particulate mass in the overflow). SE is used to evaluate particulate concentrations in the clarifier overflow (X_{OF}) and underflow (X_{UF}) by the following equations:

$$X_{UF} = \frac{SE.X_2.(R_2.q+1)}{R_2.q+q_W} \quad \text{.......} \quad (1)$$

$$X_{OF} = \frac{(1-SE).X_2.(R_2.q+1)}{q+q_W} \quad \text{.......} \quad (2)$$

where X_2 is the concentration (mg COD/l) of any particulate in the aerobic unit and its effluent.

2.2 Mathematical Model

The WWTP mathematical model follows the change in a group of variables as they pass through the system shown in Figure 1. As indicated in the BSM1 benchmark (Alex et al., 2008), this benchmark model uses the framework of the International Water Association (IWA) Activated Sludge Model 1 (ASM1) (Henze et al., 2000).

Table 1 lists these state variables with their rates; S refers to soluble concentrations and X refers to particulate concentrations.

Table 1. State variables in the benchmark model and their associated rates (Alex et al., 2008); a: Chemical Oxygen Demand.

Variable	Symbol	Rate
Soluble inert organics	S_I (mg COD[a]/l)	$r_{SI}=0$
Readily biodegradable (soluble) substrate	S_S (mg COD/l)	$r_{Ss}=\frac{-1}{y_H}(P_1+P_2)+P_7$
Particulate inert organics	X_I (mg COD/l)	$r_{XI}=0$
Slowly biodegradable (particulate) substrate	X_S (mg COD/l)	$r_{Xs}=(1-f_P)(P_4+P_5)-P_7$
Active heterotrophic biomass	X_{BH} (mg VSS COD/l)	$r_{XBH}=P_1+P_2-P_4$
Active autotrophic biomass	X_{BA} (mg VSS COD/l)	$r_{XBA}=P_3-P_5$
Non-biodegradable particulates	X_P (mg COD/l)	$r_{Xp}=f_P(P_4+P_5)$
Dissolved oxygen	S_O (mg Oxygen/l)	$r_{So}=-\frac{1-y_H}{y_H}P_1-\frac{4.57-y_A}{y_A}P_3$
Nitrate	S_{NO} (mg N/l)	$r_{SNO}=\frac{1-y_H}{2.86y_H}P_2+\frac{1}{y_A}P_3$
Free and ionized ammonia	S_{NH} (mg N/l)	$r_{SNH}=\left(-i_{XB}-\frac{1}{y_A}\right)P_3-i_{XB}(P_1+P_2)+P_6$
Soluble biodegradable organic nitrogen	S_{ND} (mg N/l)	$r_{SND}=P_8-P_6$
Particulate biodegradable organic N	X_{ND} (mg N/l)	$r_{XND}=(i_{XB}-i_{XP}f_P)(P_4+P_5)-P_8$
Alkalinity	S_{ALK} (mol/l)	$r_{SALK}=-\frac{i_{XB}}{14}P_1+\left(\frac{1-y_H}{14\times2.86\times y_H}-\frac{i_{XB}}{14}\right)P_2-\left(\frac{1}{7y_A}+\frac{i_{XB}}{14}\right)P_3+\frac{P_6}{14}$

The various processes shown in Table 1 are defined and their mathematical models, along with the rates governing all the different state variables (S and X) in the benchmark model are listed in Table 2.

Table 2. The processes found in the basic benchmark model (Alex et al., 2008).

Process	Mathematical Model	Symbol
Aerobic growth of heterotrophs	$\mu_H\frac{S_s}{S_s+K_s}\frac{S_O}{S_O+K_{OH}}X_{BH}$	P_1
Anoxic growth of heterotrophs	$\mu_H\frac{S_s}{S_s+K_s}\frac{K_{OH}}{S_O+K_{OH}}\frac{S_{NO}}{S_{NO}+K_{NO}}\eta_G X_{BH}$	P_2
Aerobic growth of autotrophs	$\mu_A\frac{S_{NH}}{S_{NH}+K_s}\frac{S_O}{S_O+K_{OA}}X_{BA}$	P_3
Decay of heterotrophs	$b_H X_{BH}$	P_4
Decay of autotrophs	$b_A X_{BA}$	P_5
Ammonification of soluble organic nitrogen	$k_a S_{ND} X_{BH}$	P_6
Hydrolysis of entrapped organics	$k_h\frac{X_s/X_{BH}}{X_s/X_{BH}+K_X}\left[\frac{K_{OH}}{S_O+K_{OH}}+\eta_h\frac{S_{NO}}{S_{NO}+K_{NO}}\frac{K_{OH}}{S_O+K_{OH}}\right]X_{BH}$	P_7
Hydrolysis of entrapped organic nitrogen	$P_7\frac{X_{ND}}{X_S}$	P_8

Definitions and values (which are used in the simulation) of the kinetic and stoichiometric parameters shown in Table 2 are listed in Table 3.

Table 3. Kinetic and stoichiometric parameters used in the simulation (Alex et al., 2008).

Parameter	Definition	Value
μ_H	Max. specific growth rate for Heterotrophs (d^{-1})	4.0
μ_A	Max. specific growth rate for Autotrophs (d^{-1})	0.5
K_S	Half saturation constant for Heterotrophs (mg COD/l)	10
K_{OH}	Half saturation constant for O_2 Heterotrophs (mg O_2/l)	0.2
K_{NO}	Half saturation constant for Heterotrophs (mg NO_3-N/l)	0.5
η_G	Correction for Anoxic Heterotrophic growth (-)	0.8
K_{OA}	Half saturation constant for O_2 Autotrophs (mg O_2/l)	0.4
K_{NH}	Half saturation constant for Autotrophic. (mg NH_3-N/l)	1.0
b_H	Decay constant for Heterotrophs (d^{-1})	0.3
b_A	Decay constant for Autotrophs (d^{-1})	0.05
k_a	Ammonification rate (l.COD/mg.d)	0.05
k_h	Max. specific Hydrolysis rate (mg COD/mg VSS COD.d)	3.0
K_X	Half saturation constant for Hydrolysis (mg COD/mg COD biomass)	0.1
η_h	Correction for Anoxic Hydrolysis (-)	0.8
y_H	Heterotrophic yield coefficient (mg VSS COD/mg COD)	0.67
y_A	Autotrophic yield coefficient (mg VSS COD/mg N)	0.24
f_p	Particulate yielding biomass fraction (-)	0.08
i_{XB}	Nitrogen fraction in biomass (mg N/mg VSS COD)	0.08
i_{XP}	Nitrogen fraction in biomass products (mg N/mg VSS COD)	0.06

Finally, Table 4 lists the characteristics of the wastewater being treated in the WWTP, which represents the flow-weighted average influent concentrations during a week of dry weather operation (Alex et al., 2008).

Table 4. Wastewater characteristics used in the simulations, which represent the flow-weighted average influent concentrations during a week of dry weather operation (Vanhooren and Nguyen, 1996).

Variable	Value
S_{Ii}	30 mg COD/l
S_{Si}	70 mg COD/l
X_{Ii}	52 mg COD/l
X_{Si}	200 mg COD/l
X_{BHi}	28 mg COD/l
X_{BAi}	0.25 mg COD/l
X_{Pi}	5.0 mg COD/l
S_{Oi}	0 mg Oxygen/ l
S_{NOi}	0.25 mg N/ l
S_{NHi}	30.0 mg N/ l
S_{NDi}	7.0 mg N/ l
X_{NDi}	11.0 mg N/ l
S_{ALKi}	10.0 mol/l

Both anoxic and aerobic units are assumed to be completely mixed, with constant liquid densities and volumes. Processes shown in Table 2 are only taking place within the units. Based on the aforementioned analysis, the following equations can be used to dynamically describe the behavior of all the soluble (S) and particulate (X) constituents in the plant; the subtext denotes the unit in the system:

For the anoxic unit (unit 1):

$$\frac{dS_1}{dt} = \frac{q}{V_1}S_i + \frac{R_1q}{V_1}S_2 + \frac{R_2q}{V_1}S_2 - \left(\frac{q}{V_1} + \frac{R_1q}{V_1} + \frac{R_2q}{V_1}\right)S_1 + r_{S1} \quad \cdots (3)$$

$$\frac{dX_1}{dt} = \frac{q}{V_1}X_i + \frac{R_1q}{V_1}X_2 + \frac{R_2q}{V_1}X_{UF} - \left(\frac{q}{V_1} + \frac{R_1q}{V_1} + \frac{R_2q}{V_1}\right)X_1 + r_{X1} \quad \cdots (4)$$

For the aerobic unit (unit 2):

$$\frac{dS_2}{dt} = \left(\frac{q}{V_2} + \frac{R_1q}{V_2} + \frac{R_2q}{V_2}\right)S_1 - \left(\frac{q}{V_2} + \frac{R_1q}{V_2} + \frac{R_2q}{V_2}\right)S_2 + r_{S2} \quad \cdots (5)$$

$$\frac{dX_2}{dt} = \left(\frac{q}{V_2} + \frac{R_1q}{V_2} + \frac{R_2q}{V_2}\right)X_1 - \left(\frac{q}{V_2} + \frac{R_1q}{V_2} + \frac{R_2q}{V_2}\right)X_2 + r_{X2} \quad \cdots (6)$$

As mentioned before, oxygen is supplied only to the aerobic unit, with the following rate (r_{Air-in}):

$$r_{Air-in} = K_L(S_O^{SAT} - S_{O2}) \quad \cdots (7)$$

where S^{SAT}_O is oxygen saturation concentration = 8 mg/l, (at 26 °C and 1 atm) (Rittmann and McCarty, 2001), and S_{O2} is the oxygen concentration in the aerobic reactor (mg/l). In order to establish the anoxic conditions, the value of this rate is zero in the anoxic unit.

2.3 Mathematical Model for QAC Fate and Effect

As discussed above, QACs negatively impact the biological processes responsible for wastewater treatment in WWTPs, with their fate and effect being connected to the processes of biological reactions' inhibition, adsorption, and their biological degradation.

QACs are mainly removed in activated sludge WWTPs through sorption and biodegradation (Ren et al., 2011; Zhang et al., 2011; Zhang et al., 2015). Sorption takes place with the biomass in the system or organic substances due to their strong affinity to organic and inorganic particles (Ismail et al., 2010; Ren et al., 2011). Biodegradation is achieved by a specific group of microorganisms during the aerobic biological wastewater treatment, with it depending on QAC concentration, structure, microbial community, and prevailing conditions (Sütterlin et al., 2008; Zhang et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015). Additionally, QACs might not be utilized by microorganisms if they are introduced at low concentrations (Hora et al., 2020). On the other hand, a high concentration will inhibit the biological reactions in the WWTP system. In general, QACs' concentrations between 10 and 40 mg/l are inhibitory to activated sludge systems (Reynolds et al., 1987), with nitrification being the most vulnerable biological process (Zhang et al., 2011).

The biodegradation of QACs will take place after an acclimation period and under conditions where readily degradable COD is somewhat limited (Zhang et al., 2011; Hajaya and Pavlostathis, 2012), both of which are not considered as instantaneous processes. Therefore, in this work, in order to reflect the scenario under consideration, the microbial community in the WWTP is assumed to be unacclimated to high concentrations of QACs, and will be affected by the processes of microbial inhibition, in addition to adsorption.

The targeted QAC compound is chosen to represent a commercially available mix that is used in a multitude of household and industrial disinfectants, which have been studied before (Hajaya et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015). The mix is comprised from three QACs (alkyl benzyl dimethyl ammonium chloride) that have a different number of carbons in their alkyl chain (12, 14, and 16).

2.3.1 QACs Inhibition

QACs' inhibition to heterotrophic bacteria responsible for COD removal and denitrification is well-documented, and can be assumed to follow the competitive inhibition model (Hajaya et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015; Conidi et al., 2019). This mood of inhibition is mathematically represented by increasing the half saturation constants for heterotrophs (K_s and K_{NO}) as a result to an increase in the inhibitory compound concentration (Rittmann and McCarty, 2001), resulting in a decreased overall reaction and growth rates:

$$\bar{K} = K(1 + \frac{I}{K_I}) \quad \text{......} \quad (8)$$

where $\bar{K}$ is the inhibited, apparent half saturation constant (mg substrate/l), K is the uninhibited half saturation constant (mg substrate/l), I is the competitive inhibitor concentration, and K_I is the competitive inhibition coefficient. Units for I and K_I are specific to the concentration of the competitive inhibitor. For COD utilization, QACs inhibit through their total concentration in the unit, while for nitrate reduction they inhibit through their soluble concentration (Hajaya and Pavlostathis, 2012). Values for the competitive inhibition coefficients are 14.9 mg QAC/l and 0.27 mg QAC/l, for COD utilization and nitrate reduction, respectively. These values are for unacclimated heterotrophic bacteria (Hajaya and Pavlostathis, 2012).

For ammonia oxidizing autotrophic bacteria, QACs inhibit the biological reactions through a non-competitive inhibition mechanism (Hajaya and Pavlostathis, 2012; Yang et al., 2015). This mood of inhibition is mathematically represented by reducing the maximum specific growth rate for autotrophs (μ_A) as a result to an increase in the inhibitory compound concentration (Rittmann and McCarty, 2001), resulting in a decreased overall reaction and growth rates:

$$\bar{\mu}_A = \frac{\mu_A}{1+\frac{I}{K_I}} \quad \text{......} \quad (9)$$

where $\bar{\mu}_A$ is the inhibited, apparent maximum specific growth rate (1/d), μ_A is the uninhibited maximum specific growth rate (1/d), I is the non-competitive inhibitor concentration, and K_I is the competitive inhibition coefficient. Units for I and K_I are specific to the concentration of the competitive inhibitor. For ammonia oxidizing autotrophic growth, QACs inhibit through their autotrophic biomass adsorbed concentration (Hajaya and Pavlostathis, 2012). Value for the non-competitive inhibition coefficients is 0.0785 mg/g VSS-COD. This value is for the unacclimated autotrophic, ammonia oxidizing bacteria (Hajaya and Pavlostathis, 2012). The susceptibility to QACs for the biological processes in the WWTP can be deduced from the value of the inhibition coefficient for each reaction; reactions with low values of inhibition coefficients will be more susceptible, affected at concentrations of the inhibitor far less that those affecting reactions with higher inhibition coefficients.

2.3.2 Adsorption

The main interaction by which QACs affect the microbial group in WWTPs is adsorption (Ismail et al., 2010; Zhang et al., 2011; Conidi et al., 2019), for its phase distribution will dictate the level of its bioavailability (and inhibition) in the system (Hajaya and Pavlostathis, 2012). The adsorption behavior of the QACs mix used in this investigation has been identified to follow the Freundlich isotherm (Hajaya et al., 2011):

$$q_e = K_F Ce^n \quad \text{......} \quad (10)$$

where q_e is QAC concentration on the biomass at equilibrium (mg/g VSS COD), Ce is QAC concentration in the liquid-phase at equilibrium (mg/l), K_F is the adsorption capacity factor (mg/g VSS COD) $(l/mg)^n$, and n is the Freundlich intensity parameter. Values for K_F and n were reported to be 11.4 (mg/g VSS COD) $(l/mg)^n$ and 0.69, respectively (Hajaya et al., 2011). QACs are assumed to undergo an instantaneous equilibrium in the WWTP units.

In the simulation, QACs' biomass association and liquid phase concentrations are followed by equating them to the total QACs' concentration. This is done by linearizing the isotherm in Eq. 10, as seen in Figure 2, and performing the subsequent mass balance.

$$q_e \cong K_P Ce \quad \text{......} \quad (11)$$

$$S_Q^T = Ce + X.q_e \quad \text{......} \quad (12)$$

$$q_e = \frac{S_Q^T}{X+\frac{1}{K_P}} \quad \text{......} \quad (13)$$

$$Ce = \frac{S_Q^T}{1+X.K_P} \quad \text{......} \quad (14)$$

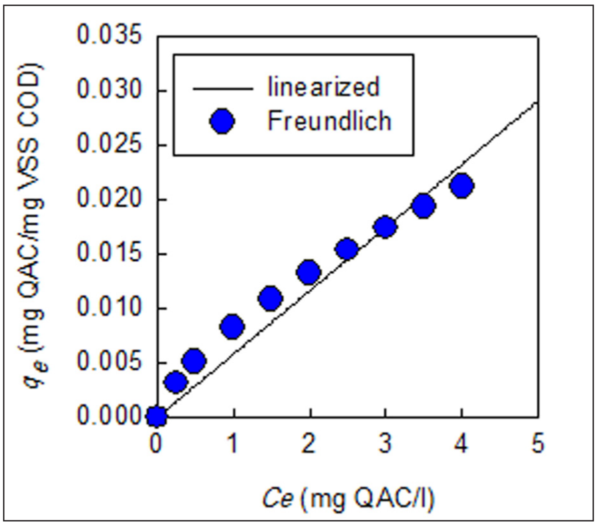

Figure 2. Linearized vs. Freundlich isotherm for the QACs mix.

where K_p is the linear adsorption coefficient for the QAC mix (mg QAC/mg VSS COD) (l/mg), S_Q^T is the total QACs concentration (mg QAC/l), and X is the total biomass available for adsorption in the WWTP (mg VSS COD/l). K_p was found to be 0.0058± 0.0002 (mg QAC/mg VSS COD) (l/mg); best estimate ± standard error. The value also considered the change in units to better suite units used in the benchmark model: (mg QAC/mg VSS COD) = (mg QAC/g VSS)/1400, 1.4 is the COD/VSS ratio (Parker et al., 2008). Furthermore, the linearization of the adsorption isotherm will result in an explicit expression for both of q_e and C_e.

2.4 Plant Simulated Operation:

The main purpose of the work presented here is to investigate the possibility of maintaining a continuous and adequate performance of a WWTP, while treating QAC-containing wastewater, by changing a group of the plant operational parameters. These changes are directed mainly to reduce the inhibitory effects by achieving an increase in microbial growth and utilization rates to overcome the reduction in their value, and/or increasing the biomass concentration in the system in order to limit the bioavailability of QACs (Conidi et al., 2019).

The WWTP depicted in Figure 1 was simulated while treating wastewater with the characteristics shown in Table 4. First, the operation treating QAC-free wastewater was simulated in order to establish the baseline performance of the plant. Second, The QAC mix was then introduced into the influent wastewater at concentrations of 2, 4, 6, 8, and 10 mg/l, individually. Plant operation was then simulated for ten days at each of the QAC concentrations in order to show the effect on the plant performance. This period of time was chosen to represent the initial response of an unacclimated WWTP to the presence of QACs at different initial concentrations. Finally, a group of operational parameters' values were changed, individually, while maintaining the others at their baseline values, in order to investigate their effectiveness in mitigating the effect of QACs on the WWTP performance.

Table 5 shows the parameters to be manipulated and the ratios and values that they will be changed to.

Table 5. Operational parameters to be manipulated and the ratios and values that they will be changed into; a: baseline operation value.

Operational parameters changed at each QAC concentration	Values changed
Solids recycle flow rate	R_2: 1[a], 1.5, 2, 4, and 6
Nitrate recycle flow rate	R_1: 1, 4, 5[a], 6, and 8
Influent wastewater flow rate	q: 0.25q, 0.5q, 0.75q, 1.0q[a], and 1.5q
Oxygen supply rate	Kla: 200[a], 230, 260, 290, 320 (1/d)

Changes in the solids' recycle ratio (R_2) were made to enhance the biomass concentration in order to limit QACs' bioavailability, while changes in the nitrate recycle (R_1) were made to enhance the growth rate of the two microbial groups in the plant. Values of the influent wastewater (q) were changed in order to limit the level of exposure to the QACs, while changes in the oxygen mass transfer coefficient (Kla) were made to increase the rate of oxygen supply to enhance the growth rate. These parameters were chosen because their alteration does not require modifying the existing layout or structure of the WWTP. The range of their values is, however, system-specific, and must be chosen within a window that insures a stable operation of the WWTP.

In order to simulate the system, the group of ODEs representing the behavior of the WWTP system was solved simultaneously in order to simulate the operation of the system. The equations were solved using the fourth-order Runge–Kutta procedure in MATLAB (The MathWorks Inc., Natick, MA), with a maximum time step of 0.5 day.

3. Results and Discussion

3.1 Baseline, QACs-Free Performance

The simulated operation for the WWTP was performed for thirty days of continuous and QAC-free, operation. Effluent steady state concentrations for S_{NH} and S_{NO} were 1.92 and 13.1 mg N/l, respectively, X_{BA} and X_{BH} of 151.2 and 2587.2 mg VSS COD/l, respectively, and the COD of 48.1 mg/l. The simulation results were comparable to the results of the benchmark study that had a similar unit sizes and feed wastewater composition and flow rate (Alex et al., 2008), and to results found in the literature for work that used the benchmark model in a wide group of applications (Sochacki et al., 2009; Ostace et al., 2011; Zeng and Liu, 2015; Harja and Naşcu, 2019; Wang et al., 2020)

3.2 Initial QACs Exposure

Figure 3 shows the effect of varying the non-ideal mixing parameters on the concentration of nitrogen compounds in the system.

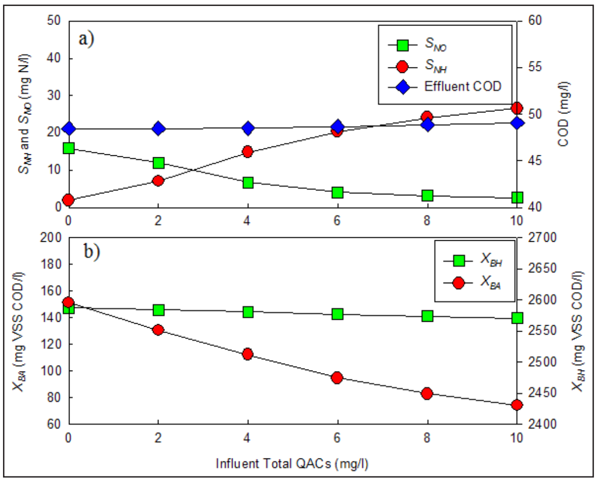

Figure 3. Effect of treating QACs-containing wastewaters at various concentrations after operating for 10 days: a) on the effluent S_{NH}, S_{NO}, and total COD, and b) on X_{BA} and X_{BH}.

As seen in the previous figure, with increasing QACs' concentration in the influent wastewater, the nitrification performance of the system has drastically dropped, resulting in an increase in S_{NH} concentration, and a decrease in S_{NO} concentration. Typically, in WWTPs, ammonia is oxidized under aerobic conditions to nitrate by autotrophic nitrifying by two consecutive biological reactions. (Madigan and Martinko, 2006; Tchobanoglous et al., 2007). However, due to QACs' inhibition, both of the biologically mediated nitrification reactions along with the microbial growth rate were inhibited. This also resulted in decreasing the autotrophic population in the system (Figure 3b).

QACs' inhibition to nitrification is well documented in the literature (Zhang et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015; Conidi et al., 2019). However, as discussed above, the WWTPs will cope with the presence of QACs in their influent wastewaters, with the effectiveness of the process reaching pre-BAC exposure levels, but not before going through an operational period that will drastically affect the plant's nitrogen removal efficiency as it is clearly seen in Figure 3a. Modifications to operational parameters should be attempted during this period in order to reduce the effect on ammonia oxidation.

On the other hand, both of the COD utilization and growth of heterotrophs were not affected in QACs at the levels found in the system feed. This can be seen clear in the relatively constant effluent COD concentration (Figure 3a) and heterotrophic biomass concentration (Figure 3b) in the system while treating QAC-containing wastewater. Similarly, this observation was also reported in the literature (Zhang et al., 2011; Hajaya and Pavlostathis, 2012; Yang et al., 2015; Conidi et al., 2019).

Therefore, the attempted mitigation actions by operational parameters' manipulations should be directed towards minimizing the effect of QACs on the nitrification process.

3.2 Possible Mitigations Actions

As discussed above, actions should be directed towards maintaining an adequate level of nitrification during the initial QACs' exposure period, in order to provide a time window for the plant to recover (through microbial acclimation, biodegradation, or adsorption), and continue its operation. The following sections discuss the various measures suggested to provide such time window, and represent the resulting difference in the performance between modified and unmodified operation while treating QAC-containing wastewater. This difference is evaluated as follows at the same QAC concentration:

$$\overbrace{\text{Difference}(\Delta)}^{\text{At the same QAC conc.}} = (S\ or\ X)^{\text{At base line parameter}}_{\text{At the same QAC conc}} - (S\ or\ X)^{\text{Changed parameter value}}_{\text{At the same QAC conc}} \quad (15)$$

3.2.1. Changing the Solids' Recycle Ratio R_2

Figure 4 shows the predicted difference in operation resulting from the simulated operation of the WWTP while treating QACs-containing wastewater at different solid recycle ratios (R_2), evaluated through Eq. 15.

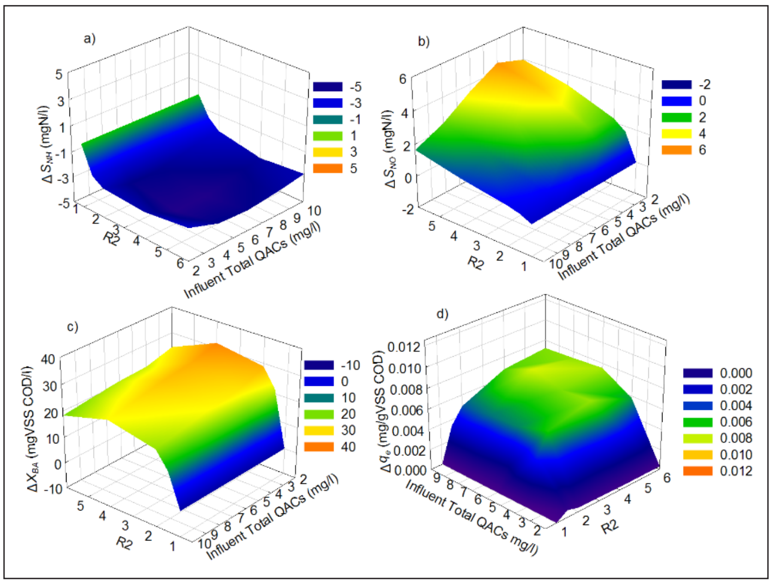

Figure 4. Difference in operation (Eq. 15) resulting from simulated operation of the WWTP while treating QACs-containing wastewater at different solids recycles ratios (R_2). A: difference in effluent S_{NH}, B: difference in effluent S_{NO}, C: difference in X_{BA}, and D: difference in q_e

As shown in Figure 4a, compared to the baseline value (R_2 = 1) increasing the solids' recycle ratio is projected to successfully reducing the effluent S_{NH} at all QAC concentrations, by up to 5 mg N/l.

This corresponded to a predictable increase in S_{NO}, less than 6 mg N/l. This more likely is attributed to a marginal enhancement in the denitrification process (Figure 4b), brought by the expected increase in heterotrophic bacteria biomass concentration of around 30 mg VSS COD/l (at R_2 = 4, at all QAC concentrations), as seen in Figure 4c. The predicted slight enhancement in Ammonia removal occurred despite the slight increase in qe, by less than 0.08 mg/g VSS COD, which is associated with the autotrophs fraction of the system's biomass (Figure 4d).

QACs are known to completely inhibit nitrification at concentrations higher than 2 mg/l (Yang et al., 2015; Conidi et al., 2019). Predicted enhancements brought by the increase of R_2 were marginal. However, they allowed for maintaining some nitrification ability and autotrophic biomass at higher concentrations (more than 2 mg/l). Additionally, this could be used in conjunction with another parameter to yield an effective mitigation measure.

3.2.2. Changing the Nitrate Recycle Ratio R_1

The predicted difference in operation resulting from the simulated operation of the WWTP while treating QACs-containing wastewater at different nitrate recycle ratios (R_1), evaluated through Eq. 1, is shown in Figure 5.

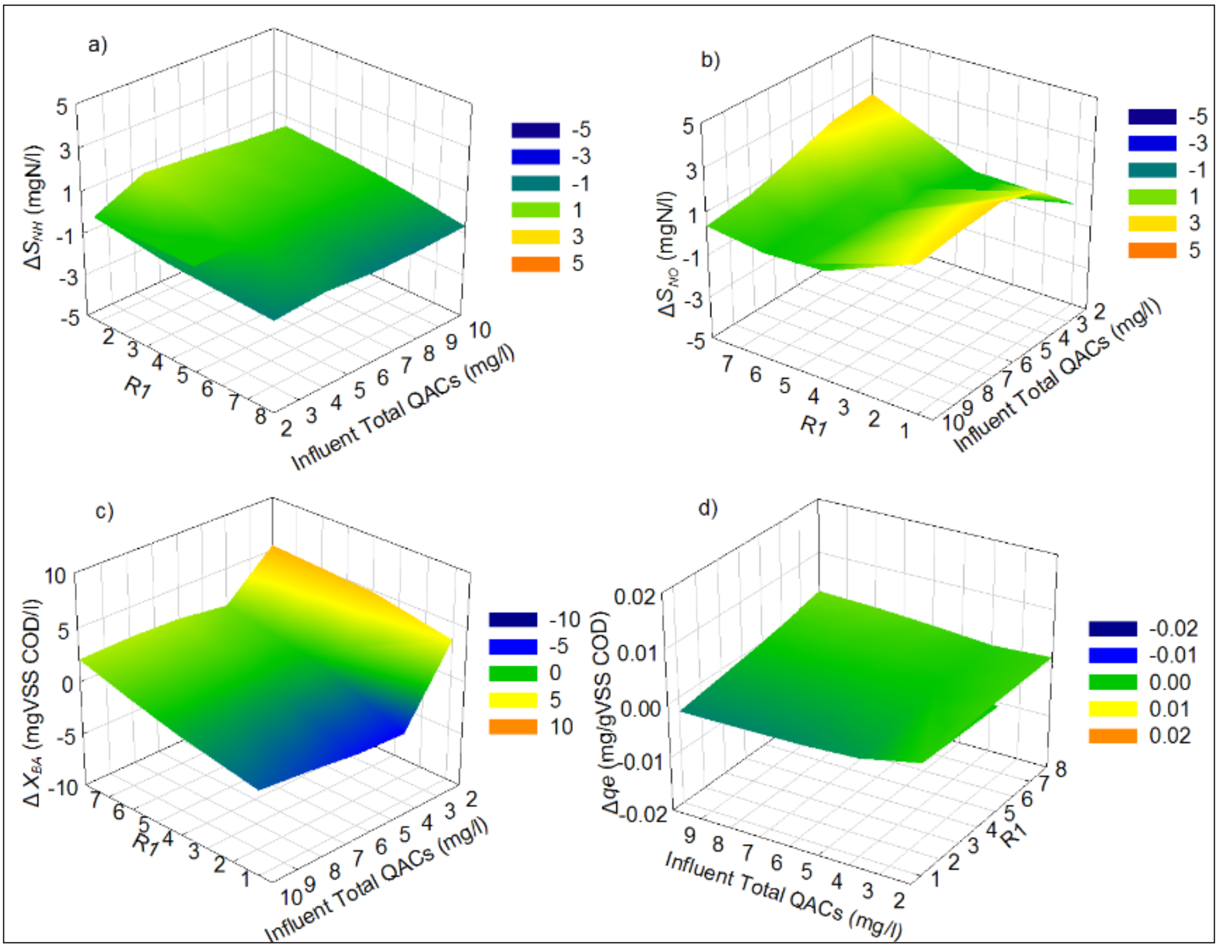

Figure 5. Difference in operation (Eq. 15) resulting from simulated operation of the WWTP while treating QACs-containing wastewater at different nitrate recycles ratios (R_1): a) difference in effluent S_{NH}, b) difference in effluent S_{NO}, c) difference in X_{BA} , and d) difference in q_e

As seen in Figure 5a, changing the nitrate recycle ratio compared to the baseline value (R_1 = 5 was not successful in improving the nitrification process, yielding slightly worse S_{NH} by almost 1 mg N/l. S_{NO} increased (by almost 5 mg N/l) when R_1 was either increased or decreased from a value of 5, as seen in Figure 5b, since this value is the best for the nitrate recycle for this system (Alex et al., 2008). Unchanged values S_{NH} were evident despite an increase in X_{BA}, up to 10 mg VSS COD/l. However, this projected increase was shown at low QAC concentrations (less than 3 mg/l), as seen in Figure 5c. Differences in qe were not projected to change and were maintained at relatively constant levels, as seen in Figure 5d. The previous projections in performance differences indicate that changing R_1 will have almost no effect in mitigating the effects of QACs on the system during its initial exposure to it.

3.2.3. Changing the Influent Flow Rate (q)

The effect of operation at different flow rate (q) fractions on the predicted difference in the simulated operation of the WWTP while treating QACs-containing wastewater (evaluated through Eq. 15) is shown in Figure 6.

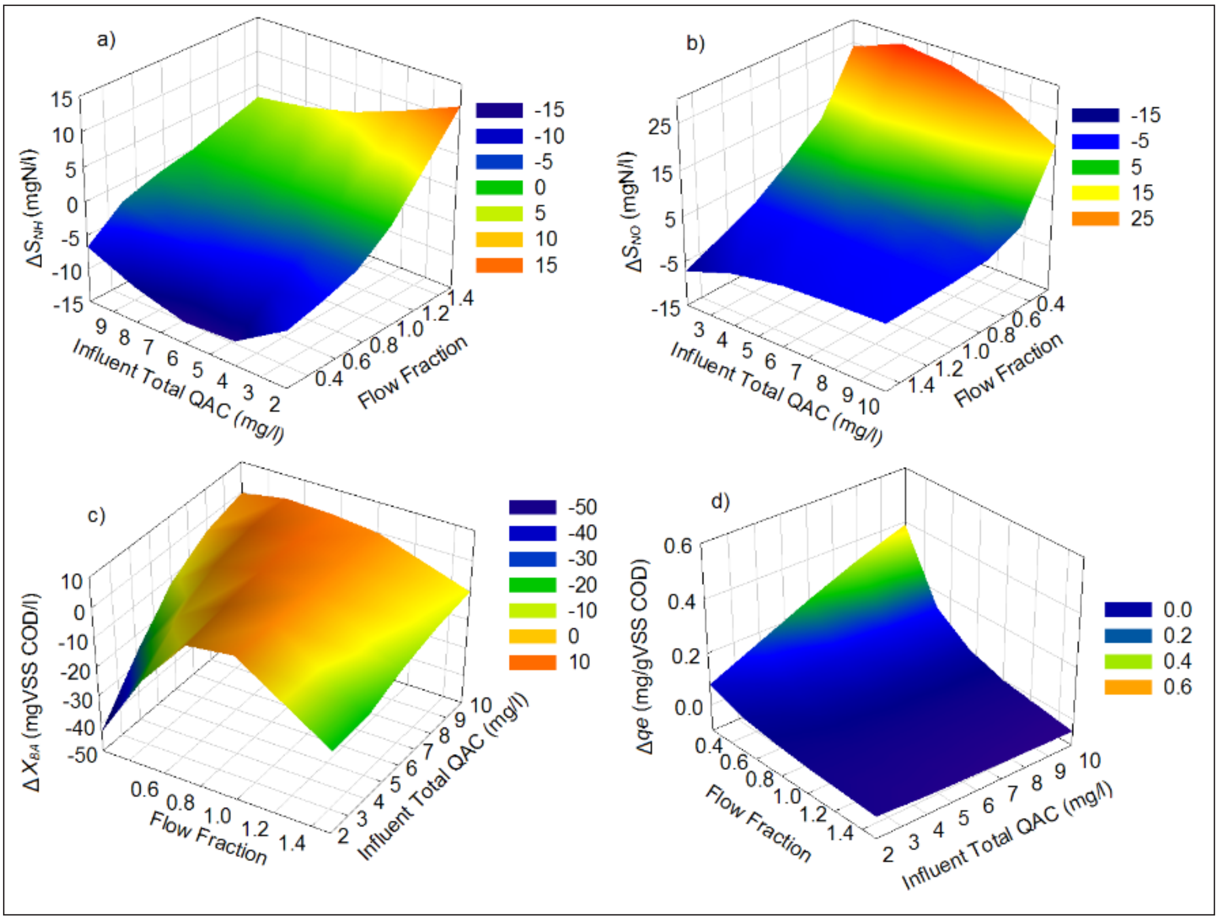

Figure 6. Difference in operation (Eq. 15) resulting from simulated operation of the WWTP while treating QACs-containing wastewater at different flow rate (q) fractions: a) difference in effluent S_{NH}, b) difference in effluent S_{NO}, c) difference in X_{BA}, and d) difference in q_e

As predicted by the simulation, increasing the influent QACs- containing wastewater did indeed reduce the nitrification performance in the system. This can be seen by increasing the effluent S_{NH}, up to almost 15 mg N/l (Figure 6a). However a decreased S_{NO} by only 5 mg N/l (Figure 6b) can be attributed to the increase in heterotrophic bacteria (from 2570 to 3320 mg VSS COD/l), which enhanced the denitrification process, as the influent wastewater flow rate is increased. This increase did not affect qe (which is associated to the autotrophs) as seen in Figure 6d. On the other hand, values of X_{BA} at the higher flow rate decreased by almost 20 mg VSS COD/l (Figure 6c). This could explain the reduction in nitrification performance.

As the flow rate was reduced, effluent S_{NH} is projected to drop up to almost 5 mg N/l (Figure 6a), which is attributed to a combination of increased of X_{BA} (up to 10 mg VSS COD/l) and reduced nitrogen load brought by a reduction in the influent wastewater. However, this enhancement is limited to drop in the feed flow rate fraction of 0.8. This reduction will also result in increasing S_{NO} by only 5 mg N/l (Figure 6b) at the same fraction limit. Changing to a smaller fraction of flow rate will eventually result in increasing S_{NO} by 25 mg N/l (Figure 6 b) , due to the decreased heterotrophic biomass concentration as a consequence of the reduced feed (from 2570 to 1667 mg VSS COD/l). qe increased drastically, between 0.2 and 0.6 mg/g VSS COD, due to the reduction in available biomass for adsorption (Figure 6d). Biomass levels in the system will affect the distribution of QACs, and ultimately affect their fate (Ren et al., 2011; Yang et al., 2015).

The aforementioned predictions for the system performance as a response to changing the influent flow rate of QAC-containing wastewater indicate that increasing the flow rate will have severe effects, similar to those resulting from extremely decreasing it (less than 0.8). The best course of action is a reduction in flow rate, for no more than a fraction of 0.8 of the original influent QACs-containing wastewater.

3.2.4. Changing the Oxygen Supply Rate

Figure 7 shows the difference in operation resulting from the simulated operation of the WWTP while treating QACs-containing wastewater at different oxygen supply rates achieved by changing the oxygen mass transfer coefficient (Kla), evaluated through Eq. 15.

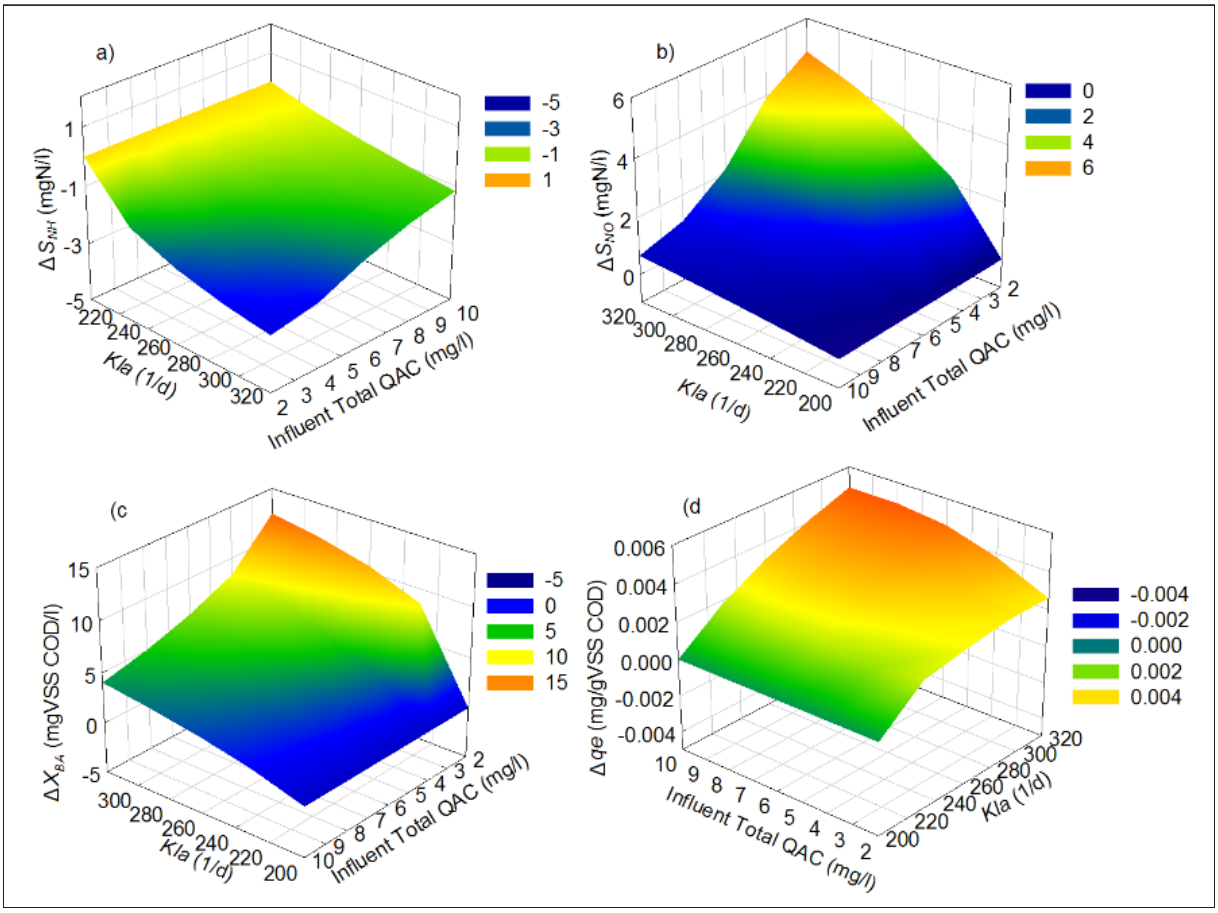

Figure 7. Difference in operation (Eq. 15) resulting from simulated operation of the WWTP while treating QACs-containing wastewater at different oxygen supply rates achieved by changing the oxygen mass transfer coefficient (Kla): a) difference in effluent S_{NH}, b) difference in effluent S_{NO}, c) difference in X_{BA}, and d) difference in q_e.

Compared to the baseline value at Kla of 200 (1/d), increasing the oxygen supply rate was capable of enhancing the system's nitrification performance by reducing S_{NH} between 3 and 5 mg N/l at Kla values between 260 and 320 (1/d), and up to a QACs' concentration of 6 mg/l (Figure 7a). The same range saw an increase in S_{NO}, between 4 and 5 mg N/l. The same could be said for X_{BA} which is projected to have an increase between 5 and 15 mg VSS COD/l (Figure 7c). S_{NO} increased (up by 6 mg N/l) at Kla values higher than 260 (1/d), remaining unchanged otherwise, as seen in Figure 7b. This is explained by the negative effect of oxygen on the denitrification process, whose rate is negatively affected by the presence of oxygen (see P2 in Table 2).

The increases in biomass also meant an increase in qe as seen in Figure 7 D. Enhancements brought by increasing the oxygen supply rate will increase oxygen concentration in the system, thus directly increasing biological reactions that utilize oxygen (Rittmann and McCarty, 2001).

On the other hand, changing Kla was not effective in mitigating the effects of QACs at concentrations higher than the aforementioned range (up to 6 mg/l). Therefore, it can be said that increasing the rate of oxygen (by changing Kla) can be used to combat the effect of QACs-containing wastewater, but to a specific limit.

4. Conclusion

The main purpose of the work presented here is to probe possible variations in a group of operational parameters of the WWTP in order to modify its operation to help reduce the initial adverse effects on the treatment, resulting from elevated QACs' concentrations in the influent. This scenario is expected as the usage of QACs-containing sanitizers has increased globally to prevent the spread of COVID-19 disease. The following conclusions were found:

The used model representing QACs' inhibition was able to successfully predict the drop in the system performance, which was mainly its nitrification ability, at each tested QACs' concentration.

Changing the solids' recycle ratio R_2 allowed for maintaining some nitrification ability and autotrophic biomass at higher concentrations (more than 2 mg/l). Best results were at $R_2 = 4$.

Manipulations in the nitrate recycle ratio (R_1) will have almost no effect on mitigating the effects of QACs on the system during initial exposure.

Increasing the influent flow rate q will negatively affect the performance, and decreasing it will help recover the performance. However, in order to maintain a stable WWTP, the reduction in q should not exceed a fraction of 0.8. This effect was predicted for QACs' concentrations below 10 mg/l.

Enhancing the oxygen transfer rate by changing Kla was effective in mitigating the effect of QACs up to a concentration of 6 mg/l. Additionally, benefits of increasing Kla were detected only at a range of 260 to 320 (1/d).

The previous predictions provided a preliminary insight into actions that can be helpful in managing the WWTP effluent during its initial QACs exposure period. As a result and as discussed in the literature, the microbial community in the plant will overcome the presence of QACs and continue a regular operation.

It must be added that this work considered changing one parameter while maintaining the others at the baseline value. Tackling the challenge by considering multiple actions towards these parameters (simultaneous change) could be more effective in achieving the main purpose, and should be considered for future works.

Finally, this work provided a framework for the simulation of WWTPs that could receive inhibitory material in their influents.

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